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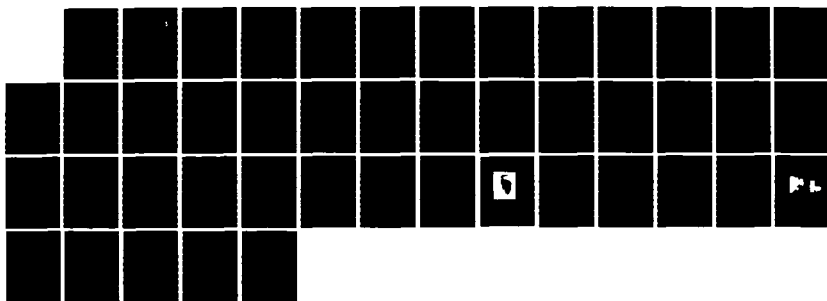
OPTICAL SPECTROSCOPY OF DYNAMICALLY COMPRESSED LIQUIDS
(U) WASHINGTON STATE UNIV PULLMAN SHOCK DYNAMICS LAB
G E DUVAL 30 JUN 86 SDL-0058-FTR N00014-77-C-0232

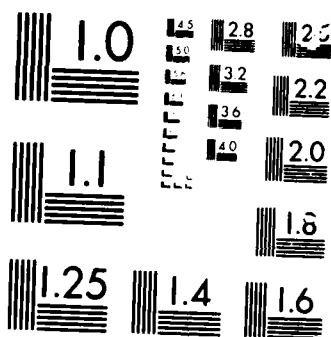
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UNCLASSIFIED

SECURITY CLASSIFICATION OF THIS PAGE

REPORT DOCUMENTATION PAGE

AD-A169 013

1b. RESTRICTIVE MARKINGS

3. DISTRIBUTION/AVAILABILITY OF REPORT
Approved for public release;
Distribution is unlimited.DTIC
ELECTE

5. MONITORING ORGANIZATION REPORT NUMBER(S)

JUL 7 1986

4. PERFORMING ORGANIZATION/REPORT NUMBER(S)

N/A since Unclassified

4. PERFORMING ORGANIZATION REPORT NUMBER(S)

SDL - 0058 FTR

6a. NAME OF PERFORMING ORGANIZATION

Washington State University
Department of Physics, SDL6b. OFFICE SYMBOL
(If applicable)

7a. NAME OF MONITORING ORGANIZATION

Office of Naval Research
University of Washington

6c. ADDRESS (City, State and ZIP Code)

Pullman, WA 99164-2814

7b. ADDRESS (City, State and ZIP Code)

3710 Brooklyn Avenue, NE, Unit No. 2
Seattle, WA 981958a. NAME OF FUNDING/SPONSORING
ORGANIZATION

Office of Naval Research

8b. OFFICE SYMBOL
(If applicable)

ONR

9. PROCUREMENT INSTRUMENT IDENTIFICATION NUMBER

N00014-77-C-0232

8c. ADDRESS (City, State and ZIP Code)

800 N Quincy Street
Arlington, VA 22217-5000

10. SOURCE OF FUNDING NOS.

PROGRAM
ELEMENT NO.PROJECT
NO.TASK
NO.WORK UNIT
NO.11. TITLE (Include Security Classification) OPTICAL SPECTROSCOPY
OF DYNAMICALLY COMPRESSED LIQUIDS (UNCLASSIFIED)

12. PERSONAL AUTHOR(S)

George E. Duvall

13a. TYPE OF REPORT

Final Report

13b. TIME COVERED

FROM 4/01/77 TO 6/30/86

14. DATE OF REPORT (Yr., Mo., Day)

June 30, 1986

15. PAGE COUNT

34

16. SUPPLEMENTARY NOTATION

17. COSATI CODES

FIELD

GROUP

SUB. GR.

18. SUBJECT TERMS (Continue on reverse if necessary and identify by block number)

Shock Waves Time-Resolved Spectroscopy CS₂
Dynamic Compression Carbon Disulfide Liquids

19. ABSTRACT (Continue on reverse if necessary and identify by block number)

The objectives of this work are described in the Introduction. Accomplishments are listed and experimental approaches are described briefly. A summary is given of spectroscopy experiments in CS₂ and other materials. Conclusions are: (i) there is no evidence in CS₂ of an unique shock effect other than that resulting from rapid increase of pressure and temperature which accompanies compression, (ii) metastable states, which can be sampled only by rapid compression, are achieved in these experiments, (iii) chemical reaction rates can be measured and critical pressures and temperatures can be determined from spectral measurements in this regime, (iv) changes in the UV and visible spectra of liquids studied vary from no observable effect to shifts of thousands of angstroms in absorption band edges for pressure less than 130 kbars, (v) observed spectral changes in liquid CS₂ are cooperative in nature and become very small or zero at infinite dilution, (vi) a combination of static high pressure and shock compression measurements is particularly valuable

20. DISTRIBUTION/AVAILABILITY OF ABSTRACT

UNCLASSIFIED/UNLIMITED ☒ SAME AS RPT ☐ DTIC USERS ☐

21. ABSTRACT SECURITY CLASSIFICATION

UNCLASSIFIED

22a. NAME OF RESPONSIBLE INDIVIDUAL

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(202) 696-4406

22c. OFFICE SYMBOL

ONR

UNCLASSIFIED

SECURITY CLASSIFICATION OF THIS PAGE

in unravelling the physical processes involved, (vii) shock experiments appear to represent a particularly useful adjunct to and extension of chemical processes in liquids under conditions of high temperatures and pressures.

OPTICAL SPECTROSCOPY OF DYNAMICALLY COMPRESSED LIQUIDS

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FINAL REPORT

4/01/77 - 6/30/86

Contract No. N00014-77C-0232



WORK SPONSORED BY THE OFFICE OF NAVAL RESEARCH

June 30, 1986

WSU Contract No. 2464-0058

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I. INTRODUCTION AND OBJECTIVES

Chemical reactions in condensed matter inevitably involve continuum mechanics and thermodynamics. When the reactions are exothermic and fast, large amplitude wave propagation is involved and the relations among the aforementioned processes are so intimate that cause and effect are difficult to separate -- passage of the shock wave initiates the reaction and the reaction drives the shock wave. Both the shock wave and the reaction influence the temperature and pressure history of a mass element participating in the reaction.

Detonation is the best known example of such a reaction. It has been studied for over a hundred years, but most effort has gone into understanding continuum mechanics aspects, with moderately sophisticated thermodynamics, primitive treatment of chemical reaction theory, and almost no attention to molecular processes. In consequence, progress in understanding the total reaction phenomenon has been slow.

During the last thirty years a substantial amount of information has accumulated on non-explosive reaction in shock waves.²⁰ This work has been done in Australia, Japan, the USA, and the Soviet Union. It suggests that the effects of a shock wave in producing chemical reactions go beyond the simple effects of pressure and temperature. This has been labeled the "catastrophic shock effect" by R.A. Graham.¹⁹ There may be direct interaction between the shock front and the individual molecules which influences their chemical states. There are certainly large numbers of structural defects produced in the shock, and these may serve as nucleation centers for initiation of reactions, or they may otherwise affect the course of the reaction.

There are also large, heterogeneous electric fields and charge distributions in insulating solids, and probably in liquids too. These may be expected to be altered in the shock front and to contribute to the total reaction process.

Plausible mechanisms of direct interaction between shock front and molecules include direct transfer of energy to particular modes, molecular deformation through impact, and torsion of large molecules. Such interactions are strongly dependent on rise time in the initiating shock, which may be the order of 10^{-12} seconds in some circumstances.

Research under this contract has addressed these problems of shock wave chemistry by means of dynamic time-resolved measurements made on samples as they were being compressed. Substances

studied have been exclusively liquid; methods have involved mechanical and electrical measurements at first and optical spectroscopy later on. The novelty of this research has required that considerable effort be directed to the development of new experimental techniques. A reasonably complete study has been conducted on carbon disulfide, and a limited survey has been made of sixteen other liquids, most of them organic. More complete statements of the shock chemistry problem will be found in references 1-4.

II. EXPERIMENTAL METHODS

Research on this contract has been sharply divided into two major, distinct phases. The first phase, consisting of mechanical and electrical measurements on shocked liquid carbon disulfide, was initiated and executed by Stephen A. Sheffield (presently at Los Alamos National Laboratory). Experimental details of his work have been reported in great detail in his thesis⁷ and in somewhat less detail in reference 15. This work provided vital information for the second phase of the work, electronic spectroscopy in the visible and ultraviolet regions. The success of the latter experiments represents the culmination of research supported under this contract, and this section will be devoted to that phase of the general problem of shock-induced reactions.

Details of the spectroscopy experiments have been described in references listed at the end of this report, particularly reference 3. The scheme of the experiments is indicated in Figures 1-4^{11,12}.

Figure 1 shows the layout of the 50 foot long gas gun which accelerates a one kilogram projectile to the velocity necessary to produce the desired pressure. The breech containing high pressure gas is at the left. The target chamber is at the right, and the sample is mounted in a target holder at the end of the barrel. The projectile and target are shown in the sketches of Figure 2. The four-inch diameter projectile has, mounted on its face, a slotted aluminum cylinder which supports a one-inch diameter sapphire impactor. At the base of the cylinder, and inside it, is a front-surfaced mirror mounted at an angle of 45° to the projectile axis. At the moment of impact of impactor and target the Xe flash lamp mounted in the target is directly opposite the slotted cylinder. Light from the lamp falls on the 45° mirror and is directed through the impactor, through the cell contained in the target, and hence to a spectrograph and streak camera. The cell consists of a thin layer of liquid encased in a cylindrical brass shell and bounded front and back by sapphire disks, F and B, as shown on the right side of the figure. The target holder is adjusted before the experiment so that the tilt between the plane faces of impactor and cell face is less than 0.5 milliradians.

The impact between impactor and the front face of the cell produces shock waves which run forward into the front cell piece and backward into the impactor. The forward-running wave induces a shock wave of pressure less than the impact pressure when it reaches the liquid sample. There follows a series of reverberations in the liquid layer between front and back sapphire disks. These eventually bring the pressure in the sample up to the impact value. The pressure buildup in a 100 micron thick sample of carbon disulfide is shown in Figure 3. It is calculated from the shock wave jump conditions, an elastic equation of state for sapphire, and an equation of state for carbon disulfide developed by S.A. Sheffield^{7,13}. The accuracy of this calculation has been confirmed in a series of experiments described in references 8 and 14. Experiments in this configuration are limited in pressure to less than approximately 130 kbars because sapphire begins to lose transparency at that pressure.

The records obtained in these experiments are illustrated in Figure 4b where time increases upward, wave length increases from right to left and transmitted light intensity is indicated by the brightness of the image.

III. ACCOMPLISHMENTS

3.1 Experimental Techniques

3.1.1 Mechanical Measurements

The development of new techniques has been a major part of this research. There have been earlier attempts to obtain transmission and reflection spectra of shock-compressed samples, but they were marginally successful at best. They did reveal some of the important difficulties of such experiments, thus making it possible to more effectively direct our efforts in this research.

The initial work by Sheffield^{7,15} involved only mechanical and electrical measurements, but it represented the first work reported on quantitative dynamic measurements in a shock-induced reaction. It involved the following novel elements:

- Cell design for liquids containing multiple EMV (particle velocity) gages.

- Use of a reflected shock to drive the sample into the reacting region with step increases in pressure and temperature.
- First observation of a double shock in CS_2 .
- First observation of a decaying precursor in a reacting material.
- First observation of quasi-periodic electrical signals associated with shock induced chemical reaction.
- First measurements of variations with pressure and temperature of chemical kinetic parameters in a shock-induced reaction.
- First observation of induction time in a shock-induced reaction.
- Use of induction times, particle velocity decay at a CS_2 sapphire interface, precursor decay, reaction wave shape, and electrical noise decay rate to obtain measures of chemical kinetic parameters.

Besides these novel developments, Sheffield also developed complete equations of state for both liquid CS_2 and its reaction products. The former, with some modifications, has stood the test of time very well. The latter has not been investigated further.

3.1.2. Spectroscopy

The development of successful spectroscopy techniques was a direct consequence of the determination and invention of K.M. Ogilvie, with liberal assistance from others in the Shock Dynamics Laboratory, particularly P.M. Bellamy.^{9,11,12} Some specific accomplishments in this aspect of the research were:

- Measurements of spontaneous light emissions from target compounds, including PMMA and "black" PMMA. Both emit light in substantial quantities when shocked, but the latter is more active than the former.¹⁶

- Use of an unshocked reflector *behind* the impactor in order to avoid degradation of the reflector surface by shock, which has been commonly experienced in attempted reflection spectroscopy.
- Development of a disposable Xe flash lamp for use in transmission experiments.
- Quantitative development of reverberating shock experiments in thin cells to obtain measures of spectral change vs. pressure and temperature.
- Development of a method for imbedding a flash lamp in the projectile for transmission spectra.
- Development of a method for mounting a flash lamp on the target for transmission spectra.
- Development of procedures for measuring the electrical resistivity of a shocked liquid in a thin cell.
- Development of a method for near-isothermal dynamic compression in a thin cell.
- Development of procedures for measuring the reversibility of shock-induced reactions when pressure is suddenly released.

Detailed descriptions of these procedures can be found in references at the end of the report, particularly references 3,7,9, and 16.

3.2 Results

3.2.1. Summary of Observations in CS₂

Most of the work done during the course of this contract has been on liquid CS₂. Fiscal year 1985 was devoted principally to exploration on other liquids, and these are best understood with CS₂ results as reference.^{11,12} A listing of experiments in CS₂ grouped according to phenomena described is contained in Table I.

The absorption spectrum of a 160 micron thick layer of CS_2 obtained with a spectrometer is shown in Figure 4a. The position of the 3500 Å absorption edge shown in the figure varies with pressure and temperature. In the experiments reported here the spectrum of CS_2 as it is shocked is recorded on film by means of spectrograph and streak camera, and variations of the band edge position are correlated with time, pressure and temperature. The records consist of photographic films in which film density varies with wave length and time while pressure changes. A copy of one of the records is shown in Figure 4b. Time in this figure increases vertically upward and wavelength increases horizontally from right to left. The band edge of interest is shown on the right at approximately 3600 Å before the shock reaches the sample. The flash lamp is turned on at the bottom of the figure, and approximately 280 nsec later at point A, the shock wave enters the CS_2 sample. When it does so, the band edge takes a step to the left by an amount approximately proportional to the pressure. As pressure in the cell rings up to its final value (as in Figure 3), the band edge steps to the left at each shock reflection. Pressure has nearly reached its final value when the band edge is at 5100 Å, indicated by the dark vertical line through the figure. Pressure remains constant during the next 250 nsec while the band edge shifts farther then regresses to a more or less constant value. At the end of the period a reflected rarefaction from the back face of the cell reaches the CS_2 and produces a rapid decrease in pressure. This is accompanied by a reversal of the band edge shift. Pressure rings down, but has not yet returned to zero when the experiment is terminated by fracture of flash lamp or sapphire.

By comparing calculated curves of pressure vs. time, as in Figure 3, with measured values of band edge position from records like that of Figure 4b, it is possible to construct the curve shown in Figure 5. The ordinate in this figure represents the shift of wave length in Angstroms produced by a given pressure, plotted as abscissa in the figure. The points shown are measured and represent five different experiments with different impact pressures and different sample thicknesses. These were selected from experiments listed in Table I-A as having the most perfect registration between steps in band edge and steps in pressure. This type of loading is denoted "SWL" or "Step Wise Loading". It refers to "thick cell" experiments, thickness being in the range 100 to 300 microns. The curve shown represents a least squares fit to the points:

$$\Delta\lambda(\text{\AA}) = 26.75p - 0.2554p^2 + 0.00225p^3 \quad (1)$$

with p in kilobars. It represents the measurements quite well, as can be seen from the figure. Some of the differences between measured and fitted values may be due to variation in impact pressure

and the resulting variation in temperature with pressure. Equation (1) should not be used for p greater than 89 kilobars because the nature of the reaction process changes at that pressure.

3.2.1.1 Reversible and Irreversible Reactions

When impact pressure is less than 89 kilobars, the reaction represented by band edge shift appears to be completely reversible. This is suggested by Figure 4b and confirmed in other experiments in which pressure falls to zero before the experiment is terminated, Table IB and Table II. There is a residual displacement of the band edge at zero pressure, but it has the magnitude expected from residual temperature which exists because of the dissipative character of shock compression. When impact pressure exceeds 89 kilobars, the band edge continues to shift toward the red after pressure has become essentially constant. The rate of shift increases monotonically with overpressure above 89 kilobars, and the band edge eventually goes off scale at about 7000 Å. This is illustrated in Figure 6.

Reference to Table IB shows how reaction rate is related to band edge reversal. Shot no. 84-045 at 89.3 kilobars shows no band edge reversal when pressure is released. No. 84-049 at 90.2 kilobars shows partial reversal. The difference is that the pressure release in 84-045 was delayed approximately 200 nsec relative to 84-049 because of the thicker back window. The curves of Figure 6 seem to show two different reaction rates. Both can be distinguished at 89.3 and 91.1 kilobars (Table IB). The second, and larger, reaction rate, expressed in Angstroms per nanosecond, is approximately 4 ($p - 88.6$) for $p > 88.6$ kilobars. The first, which is not distinguishable at 93 kbs and above, is approximately 2.5 ($p - 88.6$) Å/nsec. If pressure is released before the band edge goes off scale, the edge returns toward the blue. Its final position lies above 3500 Å by an amount which appears to be a measure of progress of the irreversible reaction.

These remarks apply to the time scale of these experiments. The results of static measurements performed by Agnew and Swanson at Los Alamos show that the static pressure threshold for irreversible reaction is about 80 kilobars at room temperature, which is lower than the value we report¹⁷ at elevated temperatures. This implies that reaction in dynamic experiments is proceeding from a metastable state, and that reaction paths may, therefore, differ from those recorded in quasi-static experiments.

3.2.1.2 Pressure and Temperature Effects

By heating or cooling the sample before it is shocked it is possible to vary the $\Delta\lambda$ vs P curve shown in Figure 5. From such measurements the effects of pressure and temperature can be separated. The results of such experiments can be accurately expressed by the following relation among band edge position (λ), pressure (P), and temperature (T):¹⁸

$$\lambda(\text{\AA}) = \sum_{n=0}^3 \sum_{m=0}^2 A_{nm} P^n X^m \quad (2)$$

where $X = T (^{\circ}K) - 295$

$$A_{nm} = \begin{pmatrix} 3550 & .379 & 7.47 \times 10^{-4} \\ 12.15 & .0492 & -4.38 \times 10^{-6} \\ -.132 & -5.11 \times 10^{-4} & 5.58 \times 10^{-7} \\ 1.69 \times 10^{-3} & 0 & 0 \end{pmatrix}$$

and P is in kilobars.

The range of validity of this expression is:

$$0 \leq P \leq 70 \text{ kb}$$

$$200 \leq T \leq 730 ^{\circ}K$$

Within this range it represents measurements with an error less than 1%. This equation also encompasses the static measurements at room temperature reported by Agnew and Swanson (op cit). The λ, P, T surface is shown in Figure 7.

3.2.1.3 Dilution

When diluted in hexane or ethanol, the band edge shift is well represented at 50 kbars, for moderate dilution, by the straight line:

$$\Delta\lambda(\text{\AA}) = 200 + 800x$$

where x is volume fraction of CS_2 , $x > 0.1$. When the CS_2 fraction is less than 10% the curve turns downward rapidly and $\Delta\lambda$ appears to vanish at infinite dilution, though it is likely that some small effect remains. These results show conclusively that the observed reactions are exclusively condensed matter reactions. Data are shown in Figure 8 and Table ID.

3.2.1.4 Thin Cell Experiments

These are listed in Table IE. The original reason for reducing sample thickness was to reduce total absorption enough that band shape could be measured. This occurred at cell thicknesses of 1 micron or less, but cooling of the CS_2 by conduction to sapphire turned out to be a problem in such cells. There is an additional reason for using thin cells, and that is to decrease the compression time. It then becomes necessary to do a series of experiments to determine how thick a cell must be if cooling effects on the spectrum are to be avoided. A 10 micron thick sample is "thick" for SWL loading; the necessary thickness for ramp loading depends on rise time of the ramp. Eight microns is thick for a 70 nsec ramp. Thirty microns is thick for a 340 nsec ramp.

An unexpected result of these experiments is that compression of a one micron thick sample with a 340 nsec ramp is essentially isothermal. That is, displacement of band edge with pressure for such an experiment is indistinguishable from values obtained by static compression at room temperature.¹⁷ This suggests the possibility for development of an isothermal, dynamic pressure gauge based on transmission spectroscopy.

3.2.1.5 Reflection

Reflected spectra show that CS_2 under the loading described here behaves like a poorly conducting metal at wavelengths shorter than that of the band edge. At longer wavelengths it is transparent, at least in the region of reversible reaction. This is illustrated in Figure 9. The reflection at 91 kb is quite intense for $\lambda < 5000 \text{ \AA}$; that at 60 kb is faint, but still discernible.

3.2.1.6 Conclusions

Band edge shift in the UV and visible is a "good" parameter for monitoring reactions, applying equally well to single shock, multiple shock, and isentropic experiments in liquids.

The observed reactions are multimolecular and do not occur at infinite dilution.

The extensive observations on CS_2 are commensurate with a process wherein rapid compression produces, reversibly, platelets of an electrically conducting substance which are a few microns in diameter and a few tenths of a micron thick.

Topological constraints, coupled with the above observations, imply that there are at least three identifiable reaction stages in CS_2 : the reversible stage described above, a second reversible stage in the pressure range 89-91 kilobars which absorbs weakly at wavelengths longer than 5000 Angstroms; and a third stage which is irreversible and which progresses at a rate that increases rapidly with pressure at pressures above the 89 kilobar critical pressure. Taken together, the reflection and thin cell experiments suggest that the first reaction produces a conducting polymer.

3.2.2 Observations on Other Liquids

The principal reasons for diversion of research effort from CS_2 to other liquids during the last year of this contract were to determine whether or not the spectral changes observed in CS_2 are unique to that material and to provide some guidance to other compounds which may also undergo major changes as a result of stepwise shock loading. A secondary objective has been to attempt to choose materials whose response to stepwise loading may provide some clue to the microscopic chemical processes which are occurring. Compounds examined have been almost exclusively organic. Stable and metastable compounds have been included. Materials and results are summarized in Table II.

The entries in Table II are, column by column:

1. Shot number.
2. Sample material.
3. Sample thickness in microns.
4. Impact pressure in kilobars.
5. Statement of pressure reversal. This is achieved by using a thin sapphire back piece to the cell and allowing the rarefaction from its free surface to return to quench the sample.

6. This is the apparent band edge before impact shown in the spectrograms.
7. $\Delta\lambda$ is the amount by which band edge is shifted toward the red when pressure has rung up to 95% of the impact value. When \geq appears before the number, it implies that the number is the recorded shift, but the preshock absorption edge was off scale in the UV.
8. $(\Delta\lambda/\Delta t)_p$ is the rate of increase of band edge wave length after pressure levels off. Here $\Delta\lambda$ is the shift toward the red which occurs *after* impact pressure is reached by ring up. Δt is the time it takes for this shift to occur. Their ratio is assumed to be a measure of the reaction rate.

3.2.2.1 Results

Table IIA: Pure nitromethane gives no indication of irreversible reaction at 125 kilobars, even when initially heated to 125°C. Addition of a small amount of ethyl diamine produces an observable reaction at 65 kilobars and a very fast reaction at 100 kilobars.

The solvents hexane, ethanol and CCl_4 all appear to be totally inert at 110-120 kbars. Acetone shows a small effect and ethyl iodide a much larger one. In neither case is there evidence of a continuing reaction at constant pressure.

Table IIB: Styrene shows a remarkable sequence of progressing reaction, presumably polymerization, as pressure is increased, with the reaction irreversible at 107 kilobars. At 65 kilobars it appears to be completely reversible, without even a residual temperature displacement. Neither dichloroethylene nor 1-hexane shows significant excitation. Acrylonitrile reacts very, very fast.

Table IIC: 1,5-hexadiene shows little, if any response. 2,4-hexadiene exhibits a large shift followed by an interval in which band edge wavelength oscillates in time.

Table IID: 2-butyne, surprisingly, shows no band edge shift whatsoever at 100 kilobars, in spite of the triple carbon bond.

Table IIE: Both benzene and nitrobenzene produce records like those found in CS_2 below the reaction threshold, suggesting that higher pressures would produce irreversible reactions. This is particularly interesting because of observations of polymerization of benzene under static conditions.

Table IIF: Two experiments in water display broad band extinction at pressures between 90 and 115 kilobars, but transmission is unaffected at higher and lower pressures. This could result from the locus of p - T states passing through the corner of a region of a different phase, or it could be an artifact of the experiment.

Hydrogen iodide responds dramatically to compression. The first shock, about 10 kilobars, produces a noticeable absorption in the blue and blue green, as might be expected from formation of free iodine. The second shock produces an immediate and total extinction of all light across the entire observable spectrum.

3.2.2.2 Discussion

The materials listed in Table II have not been investigated in depth, as has CS_2 , and the indicated response to compression should be taken as preliminary only. In general, it appears that inorganics with unsaturated bonds are slightly more apt to react than those with saturated bonds, and this result is in accord with meager static data that is available. It would be helpful to be able to go to higher pressures, but to do so would require extensive investigation of optical properties of components at higher pressures.

Promising candidates for more detailed study at pressures below 130 kilobars are sensitized nitromethane, styrene, acrylonitrile, 2,4-hexadiene, water, and hydrogen iodide. They are certainly interesting, but unless further data becomes available, it is hardly profitable to speculate about the processes which are occurring. A more complete discussion of these experiments will be found in reference 6.

IV. CONCLUSIONS

This work has shown that it is possible to make dynamic measurements in chemically reacting, shock compressed systems, including spectral absorption. The spectral effects of compression in some substances are very large, dramatic, highly reproducible, and unexplained. It is evident that in the case of CS_2 , reversible and irreversible reactions are generated and that the latter start from metastable states not reached in static experiments. Other than this metastable aspect and rapid response there is no indication that parameters other than pressure and temperature are involved in the reactions. No "catastrophic" shock effect has been detected.

Comparisons with static measurements suggest that the phenomena in CS_2 , at least, occupy a scientific niche which is contiguous to that of quasi-equilibrium condensed matter chemistry, but sufficiently different to provide a rich field for future research and expansion or modification of conventional concepts.

V. PAPERS AND PUBLICATIONS

5.1 Oral Presentations

"Introduction to the Physics and Chemistry of Fast Reactions," Workshop on Fundamental Research Directions for Decomposition of Energetic Materials, Berkeley, CA, January 20-21, 1981. Sponsored by Office of Naval Research, Air Force Office of Scientific Research, and Army Research Office.

"An Assessment of the Contributions of Microscopic Processes in the Shock Front to Shock-Induced Chemical Reactions," Workshop on Shock Chemistry, Palo Alto, CA, June 22, 1981. Sponsored by Sandia National Laboratories, The Office of Naval Research, and the Defense Advanced Project Agency.

"Electronic Spectroscopy in the Shock Environment," Workshop on Shock-Induced Chemistry, SRI International, Menlo Park, CA, February 2-3, 1982. Sponsored by SRI International and Office of Naval Research.

"Shock-Induced Reactions in Liquid CS₂: Density and Spectral Changes in the UV and Visible," Workshop on Initiation of Chemical Decomposition of Energetic Materials, Great Oak Landing, Chestertown, MD, May 4-6, 1982. Sponsored by Office of Naval Research.

"Effects of Temperature on the Spectrum of CS₂ in Isentropic and Isothermal Dynamic Loading," Workshop on the Initiation of Chemical Decomposition of Energetic Materials, Great Oak Landing, Chestertown, MD, August 15-17. Sponsored by Office of Naval Research.

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5.4 Dissertations

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Richard H. Granholm, "Temperature and Pressure Components of Shock-Induced Changes in the Electronic Spectrum of Carbon Disulfide," thesis submitted in partial fulfillment of the requirements for the degree of Master of Science in Chemistry, Washington State University, Department of Chemistry, August 1985.

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9. "Time Resolved Spectroscopy of Shock-Compressed Liquid Carbon Disulfide," Kendal M. Ogilvie, Jr., dissertation submitted in partial fulfillment of the requirements for the degree of Doctor of Philosophy, Washington State University, Department of Physics, 1982.*
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Table I. Summary of CS₂ Experiments

A. Standard Thick Cell Experiments

Shot Number	Cell Thickness, microns	Projectile Velocity, mm/ μ sec	Impact Pressure, kilobars	λ_0^* , Å	Grating Lines/mm	Writing Speed, mm/ μ sec
82-015	180	.323	73.1	3560	600	8.247
82-018	157	.410	93.1	3560	150	8.68 - 8.74
82-028	116 \pm 4	.342	77.4	3590	150	8.45
82-031	165 \pm 5	.246	55.4	3630	150	8.428
82-037	160	.369	83.6	3620	150	8.5 - 8.6
82-039	152	.538	122.8	3630	150	8.716

* Band edge measured on film before shock arrival.

B. Reversibility and Chemical Kinetics

Shot Number	Cell Thickness, microns	Projectile Velocity, mm/ μ sec	Impact Pressure, kilobars	Back Window, mm	Pressure Reversal	Wave Length Reversal	Reaction rate*	
							(1)	(2)
84-006	320	.35	76.7	2	yes	yes	--	--
84-025	102	.34	76.9	2	yes	yes	--	--
84-036	81	.386	87.4	1	yes	yes	--	--
84-046	25.5	.3909	88.6	11.3	no	--	2.2	--
84-045	18	.394	89.3	3	yes	no	2.4	6.44
84-049	21	.3983	90.2	2	yes	partial	--	--
84-044	19.3	.4018	91.1	3	yes	no	5.3	23.2
84-039	21	.411	93.2	3	yes	no	6.6	11.42
84-051	26	.415	94.1	2	yes	no+	--	--
84-028	43	.4345	95.6	3	yes	no	--	27.1

* There are large uncertainties in these values.

+ Band edge goes off scale and does not reappear when pressure reverses.

C. Effects of Changing Initial Temperature on Band Edge Position

Shot Number	Cell Thickness, microns	Projectile Velocity, mm/ μ sec	Impact Pressure, kilobars	λ_0 , Å	Grating lines/mm	Initial Cell Temperature, °C
83-022	174	0.298	67.4	3680	150	125
83031	151	0.281	63.4	3575	150	-72

D. Dilution of CS₂

Shot Number	Cell Thickness, microns	Projectile Velocity, mm/μsec	Impact Pressure, kilobars	Solvent	CS ₂ Concentration, Vol. %	Δλ at 50 kbar, Å
83-015	155	.327	74.0	ethanol	25	390
84-014	310	.3376	74	hexane	37	493
84-017	163	.3385	74.3	"	10	293
84-026	157	.345	75.6	"	1	16
84-031	315±3	.32	70.1	"	3	134
85-043	170	.317	69.4	"	80	835

E. Thin Cell Studies
(Cooling of sample by conduction)

Shot Number	Cell Thickness, microns	Projectile Velocity, mm/μsec	Impact Pressure, kilobars	λ_0 , Å	Grating lines/mm	Writing Speed, mm/μsec	Effects of Cooling on band edge shift
83-040	0.6	.420	92.3	3285	150	8.386	Very large reduction in Δλ
85-034	0.8	0.257	55.3	3240	150	8.488	" " "
83-037	0.9	0.368	83.4	3370	150	8.459	" " "
85-052	1.2	0.266	57.5	—	300	—	" " "
83-041	5	0.420	92.3	3430	150	8.430	Some reduction in Δλ
83-049	10	0.417	91.7	3470	150	8.486	No reduction in Δλ
Ramp Compression							
83-021*	1.0	0.447	40.5	3330	150	8.543	(338 nsec rise time) - isothermal
83-042*	4	.449	41.2	3370	150	8.595	- 50% reduction in Δλ
82-006	8	.240	35	3390	300	8.7	(70 nsec rise time) no apparent effect
84-043*	30	.446	41	3440	150	8.53	(340 nsec rise time) no cooling effect

* Fused quartz impactors. All others were sapphire.

Table II. Other Materials

I. Organic Molecules

Shot No.	Liquid	Cell Thickness (microns)	Pressure (kilobars)	Pressure Reversal	$\lambda_0, \text{\AA}$	$\Delta\lambda, \text{\AA}$	$(\Delta\lambda/\Delta t)_p$ $\text{\AA}/\text{nsec}$
A. No Carbon Double Bonds							
82-036	Pure NM	107	122.6	No	3140	314	0
84-038	" ϕ 125 deg C	137	125	No	—	200	0
85-007	NM/5wt% ED	310	65	No	3354	850	1.1
85-026	" "	122	86	Yes	3264	810	2.6
85-004	NM/10wt% EDA	160	100	No	2900	>3100	>100
85-002	" "	30	133	No	2900	>3200	>100
85-016	ethanol	110	116	No	2670	<220	—
84-009	n-hexane	160	123	No	—	0	0
85-008	carbon tet	112	111	No	2700	0	0
85-006	acetone	109	111	No	3030	70	0
85-023	ethyl iodide	158	101	No	3170	540	0
B. 1 Carbon Double Bond							
85-022	Styrene	89	58	Yes	3000	335	0.25 ^h
85-021	" "	94	79	Yes	3010	450	0.41 ⁱ
85-009	" " (p)	109	110	No	2980	900	0.55
85-017	" "	107	111	Yes ^a	3040	920	1.7
85-014	" "	107	123	No	2950	1125	0.7
85-028	cis 1,2 - dichloroethylene	156	101	No	2690	≥ 255	0
85-024	1 - hexene	158	101	No	2875	<775	-
85-010	acrylonitrile	104	110	No	2940	m	>100
C. 2 Carbon Double Bonds							
85-030	1,5 - hexadiene	155	101	No	2880	<530	-
85-041	" "	157.5	129	No	"	$\geq 710^j$	-
85-031	2,4 - hexadiene	160	130	No	2775	670	n
D. Triple Carbon Bonds							
85-027	2 - butyne	155	101	No	3260	k	-
E. Benzene Derivatives							
84-053	benzene	117	112	Yes ^k	2695	411	0
84-052	" "	305	123	No	2693	525	- ^l
85-003	nitro benzene	112	135	No	3955	≥ 870	0
	vinyl benzene (styrene)			see (B) above			

Table II. Other Materials, cont'd

II. Inorganic Molecules

Shot No.	Liquid	Cell Thickness (microns)	Pressure (kilobars)	Pressure Reversal	$\lambda_0, \text{\AA}$	$\Delta\lambda, \text{\AA}$	$(\Delta\lambda/\Delta t)_b$ $\text{\AA}/\text{nsec}$
84-040	Water	308	125	No	3260 ^b	- ^c	0
84-048	" "	310	123	No	"	"	"
85-033	Hydrogen iodide	161	120	No	3475	315-1600	(e)

- a. Early light loss, but band edge reversal is indicated.
- b. Cutoff at this wavelength is apparently due to impurities.
- c. Total extraction between about 90 and 110 kbars.
- e. The first shock in HI caused increased absorption in the red. The second shock produced immediate and total opacity.
- f. Abnormal termination of record - probably due to faulty sapphire.
- g. Band edge at 2809 \AA after pressure reverted to zero.
- h. Band edge is displaced upward approximately 13 \AA after pressure returned to zero.
- i. Band edge is displaced upward approximately 240 \AA after pressure returned to zero.
- j. Unusual features in this record suggest that there may have been artifacts from faulty sapphire.
- k. No change observed.
- m. Immediate reaction produced total opacity.
- n. Band edge position oscillated after impact pressure was reached.
- p. Inhibitor was removed by distillation.

NM denotes nitromethane.

EDA denotes ethyl diamine.

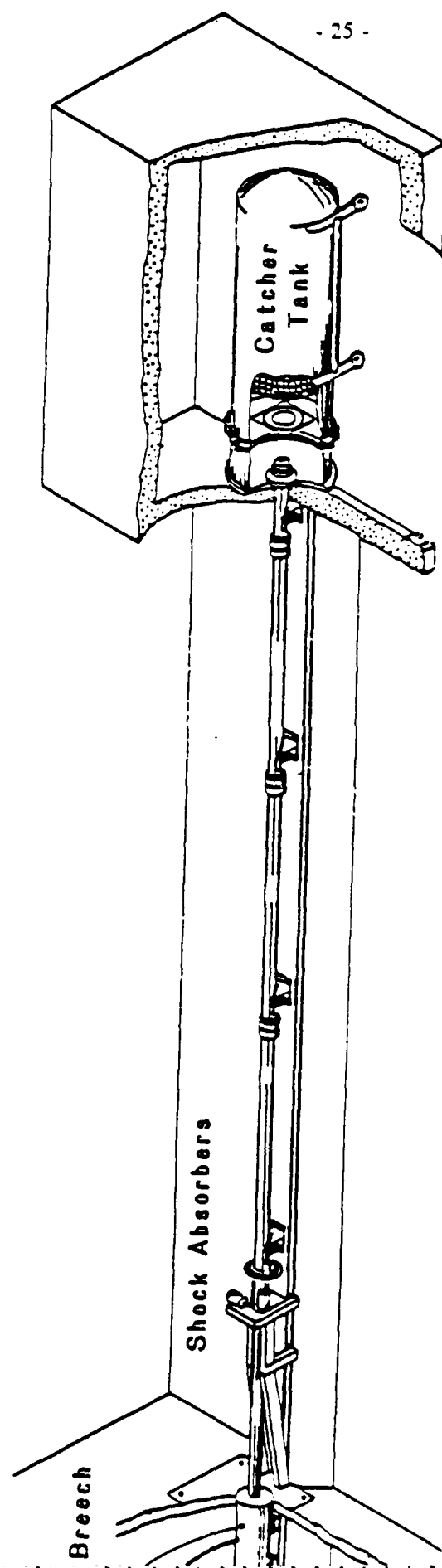


Fig. 1. Schematic diagram of single stage gas gun used to accelerate projectile. Target holder is mounted on the wall of the chamber at the right.

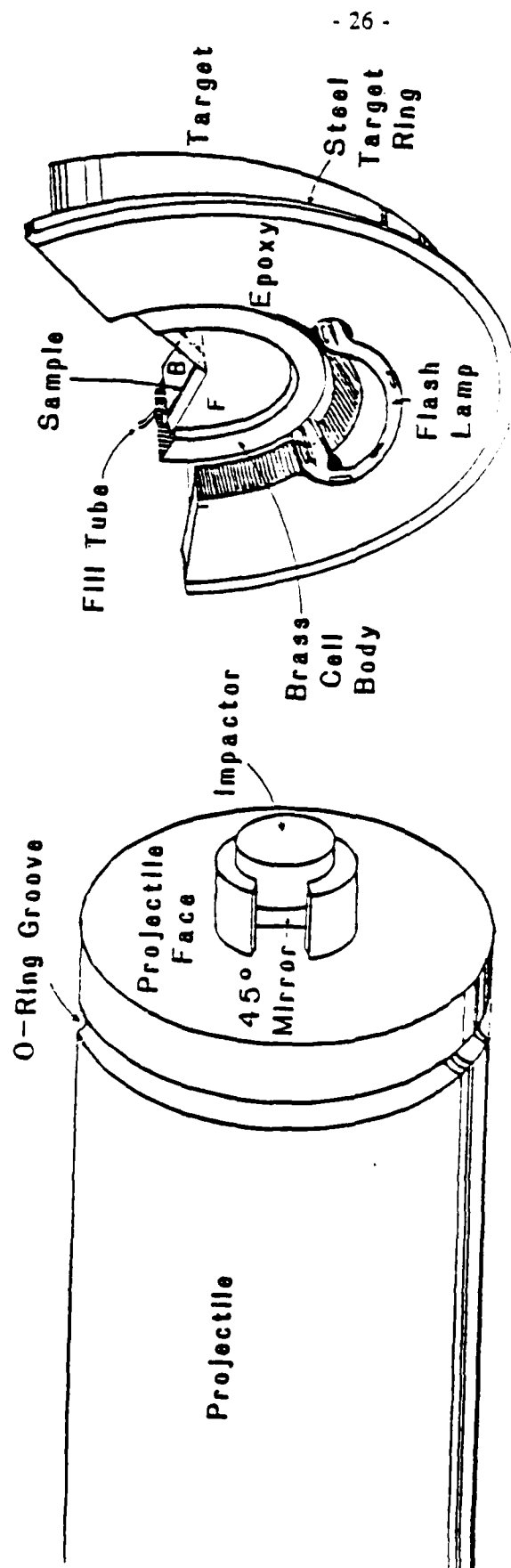


Fig. 2. Sketch of projectile face and target. Light from the flash lamp is intercepted by the 45° mirror on the projectile and turned so it passes through impactor and sample.

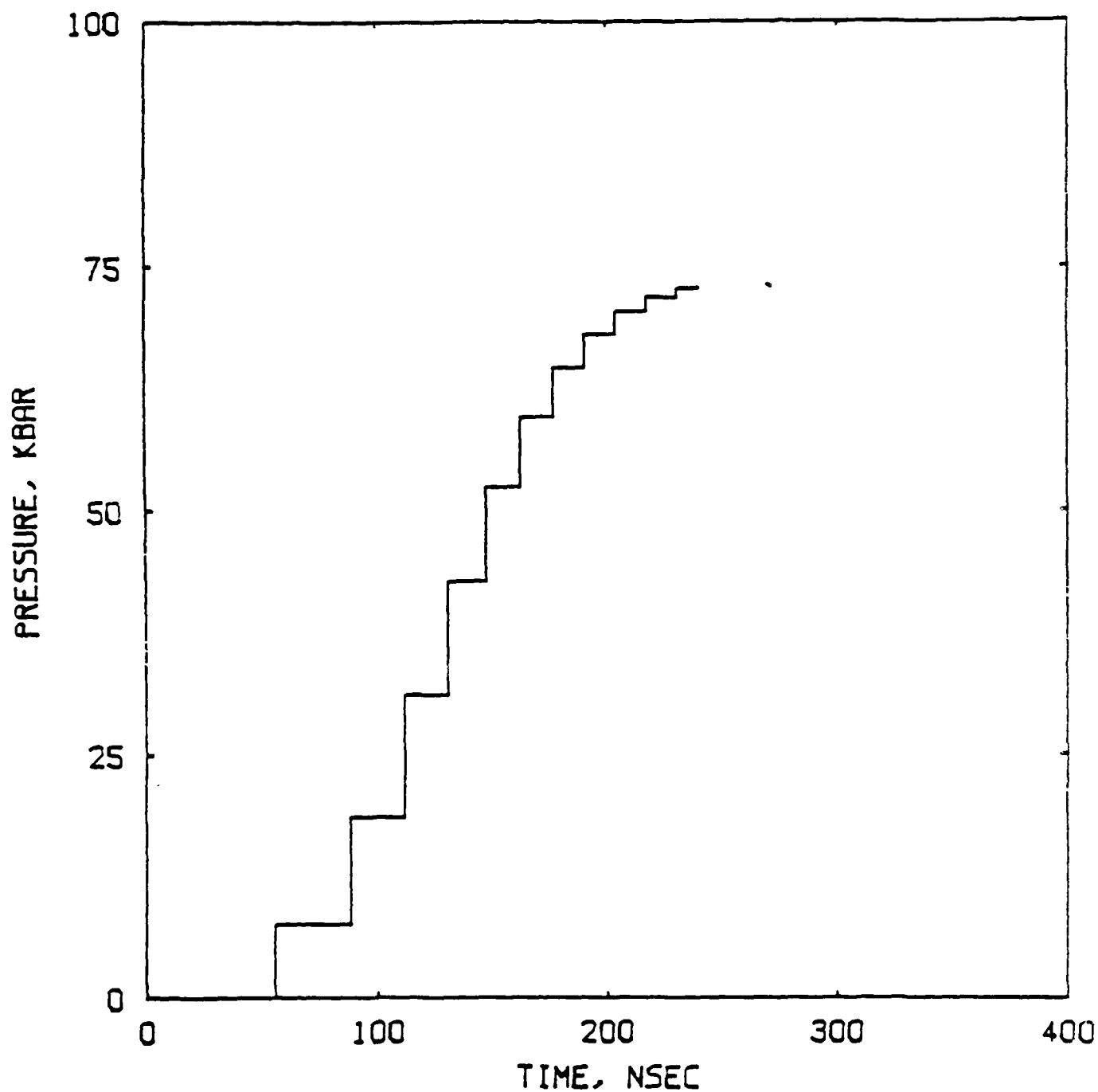


Fig. 3. Pressure in a 100 micron thick CS_2 sample between sapphire plates calculated from shock jump conditions and the SheffieldII equation of state for CS_2^{13} .

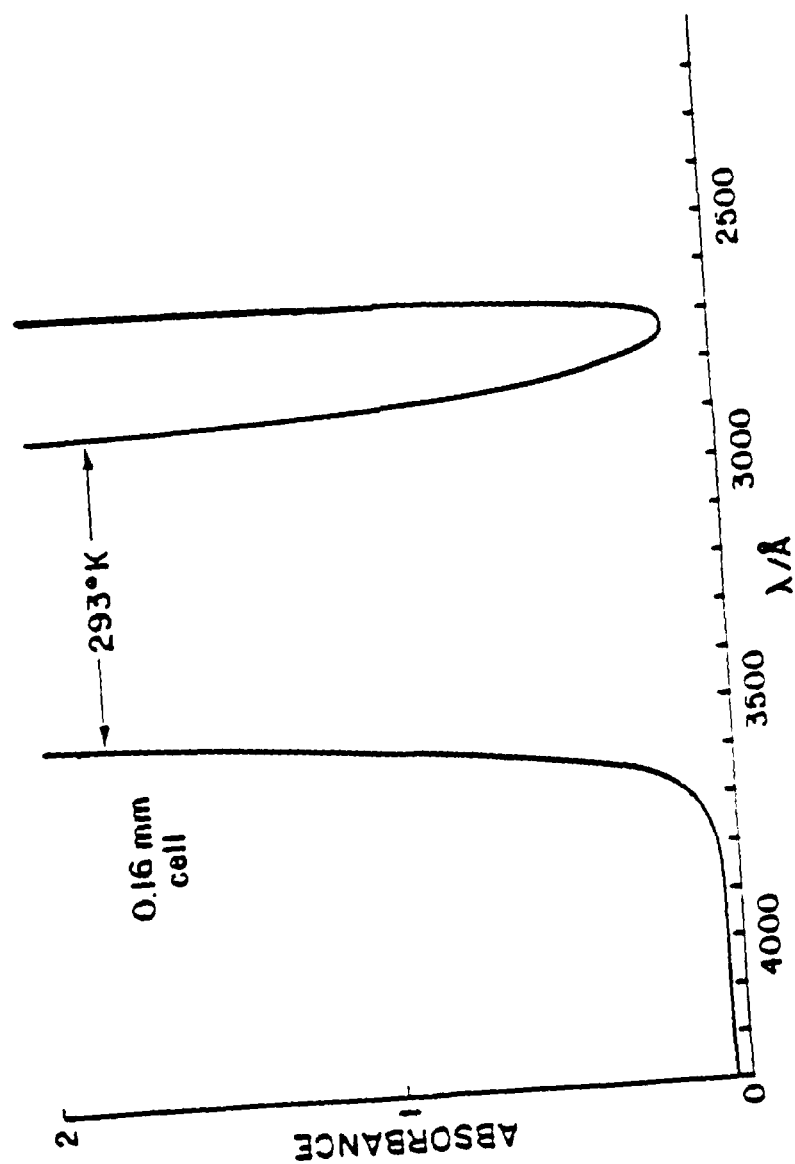


Fig. 4a. Absorption spectrum of a 0.16 mm layer of liquid CS_2 measured in a spectrometer.

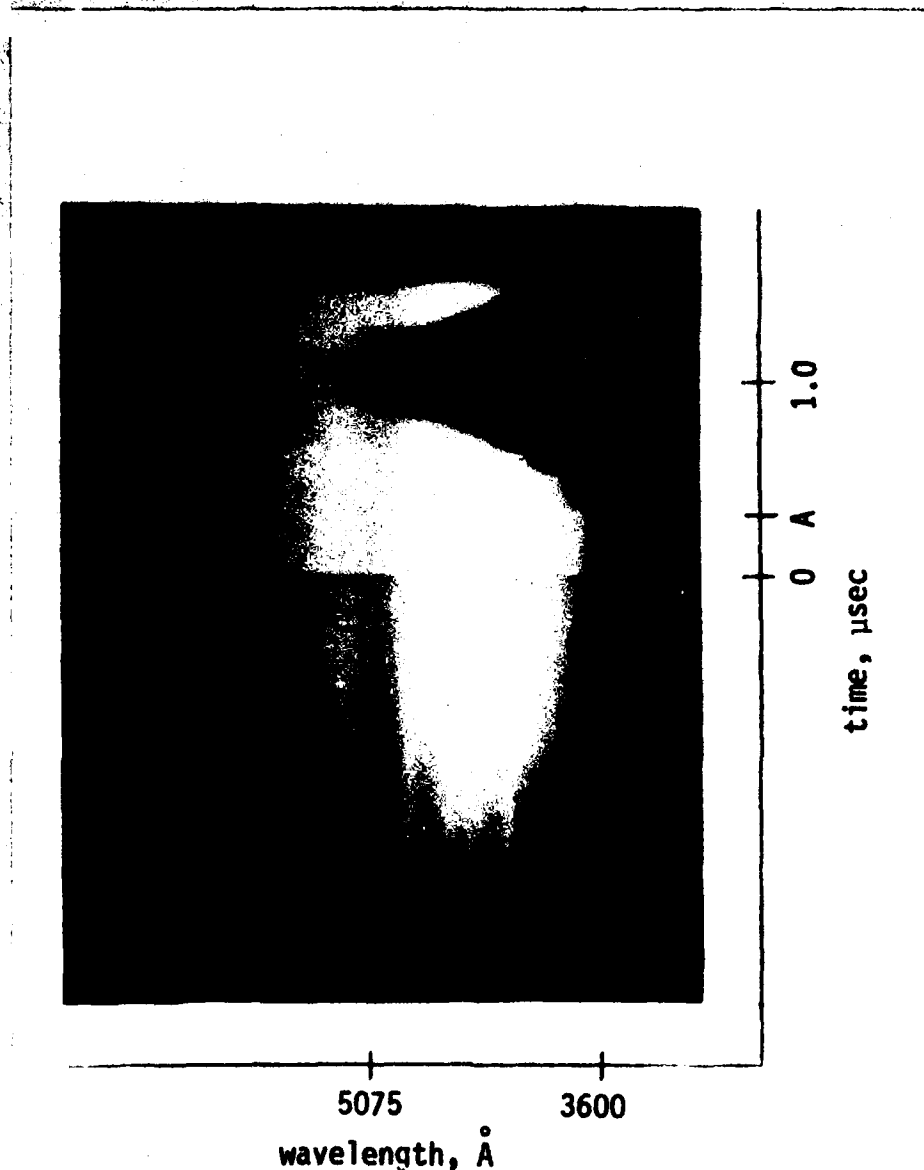


Fig. 4b. Absorption spectrum of a 0.32 mm layer of CS₂ between two sapphire plates shocked to 77 kbar (84-006). Time increases from bottom to top; wavelength increases from right to left. Impact occurs at $t = 0$.

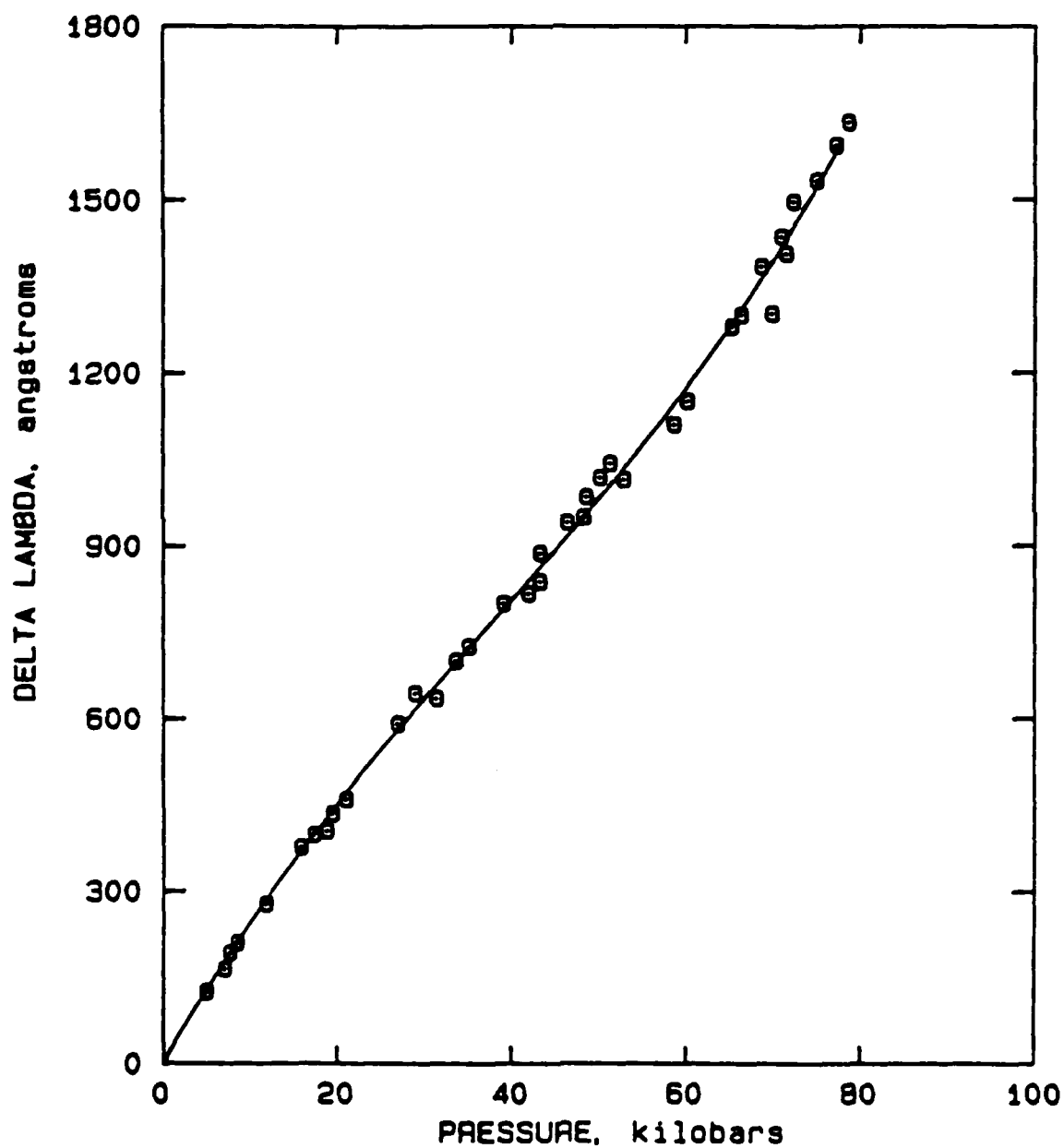


Fig. 5. Shift of red band edge due to stepwise shock loading. Reference edge is approximately 3550 Å. Points are the result of five experiments. The line is $\Delta\lambda = 26.75p - 0.2554p^2 + 0.002252p^3$, p in kbar, $\Delta\lambda$ in Å. Samples were at room temperature before they were shocked.

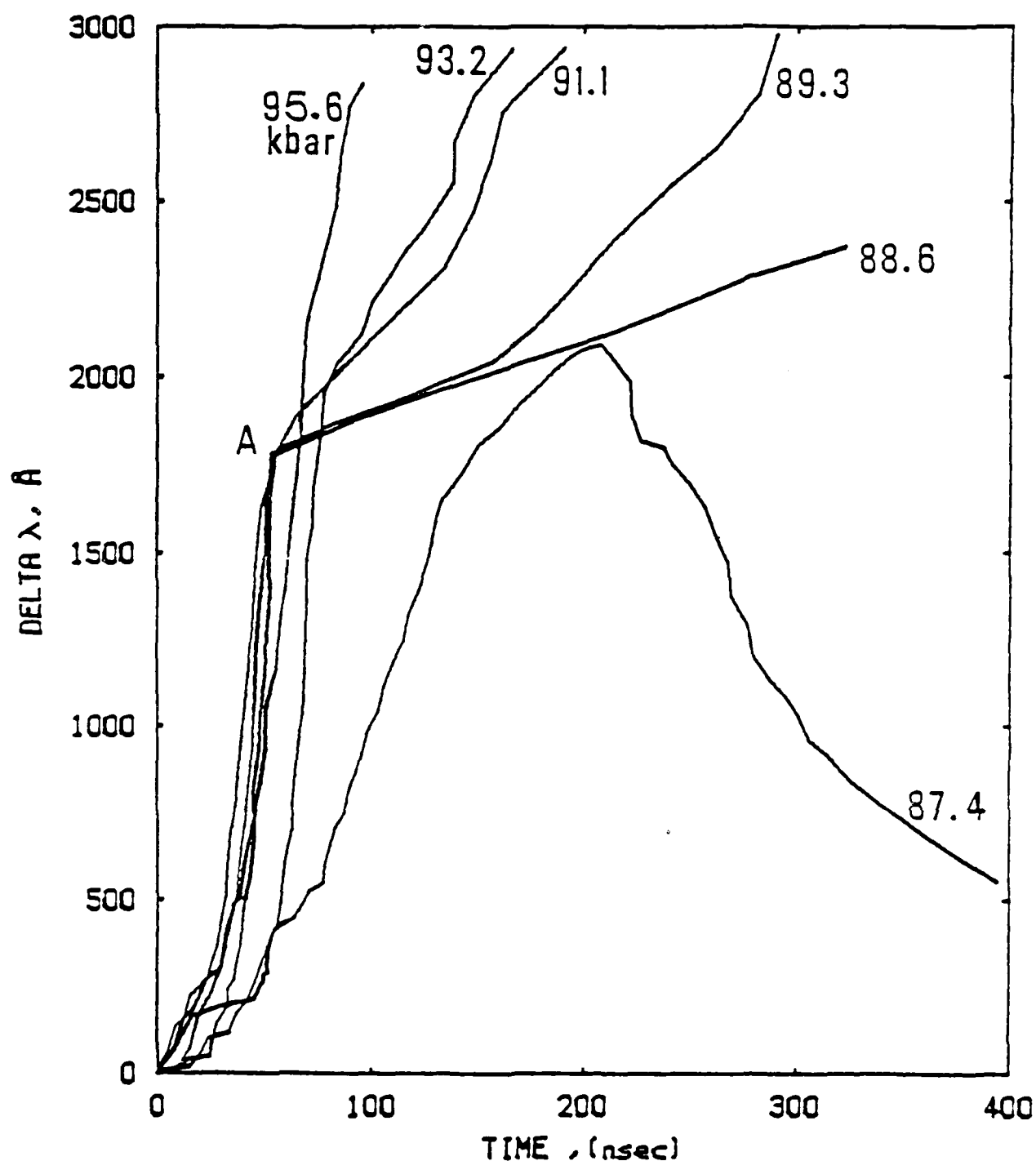


Fig. 6. Reversibility threshold and reaction kinetics for CS_2 shocked to different pressures (numbers associated with the individual curves are impact pressures in kbars).

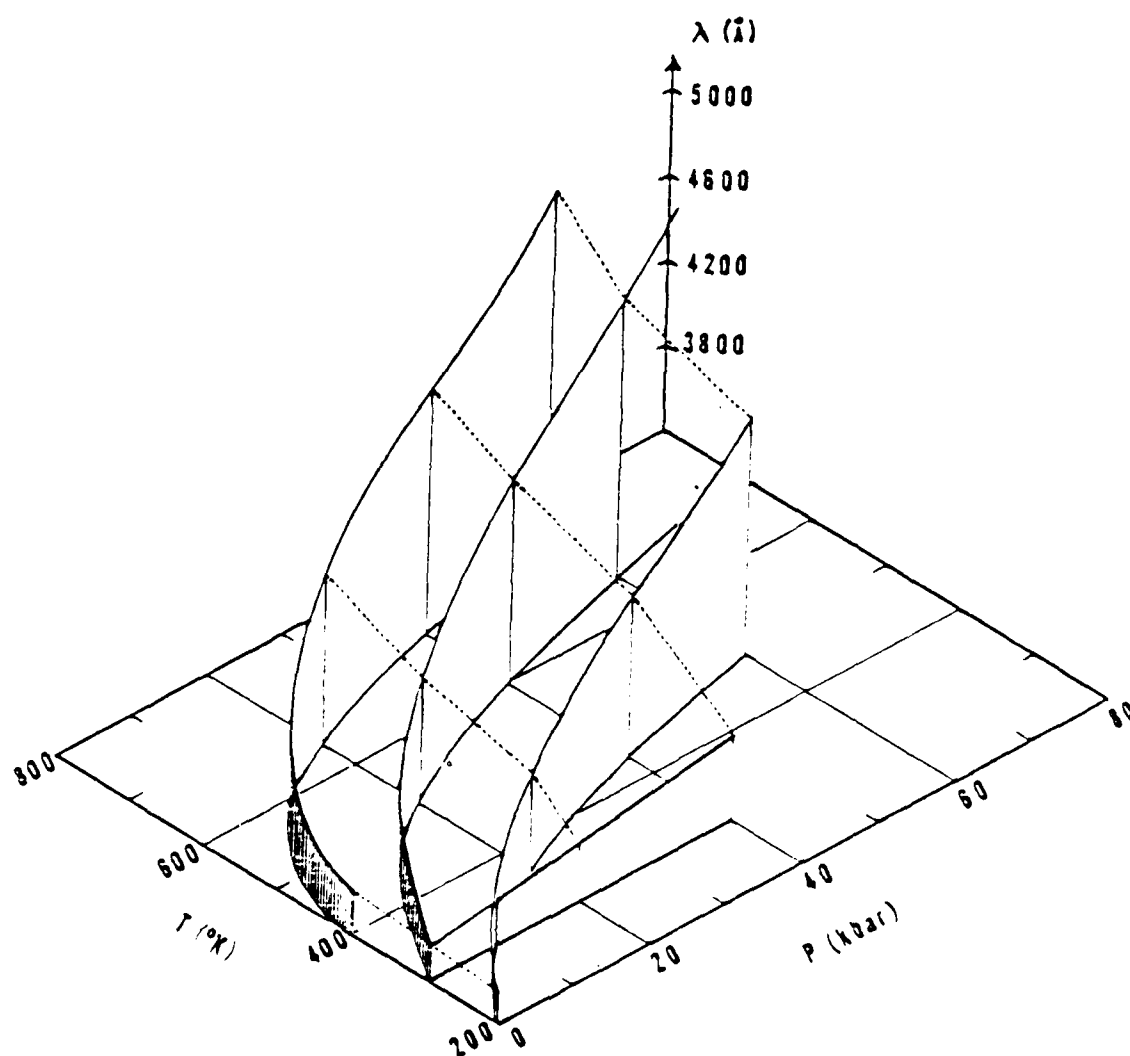


Fig. 7. Measured wavelengths of 3200 \AA band edge from hot, cold, and room temperature experiments.

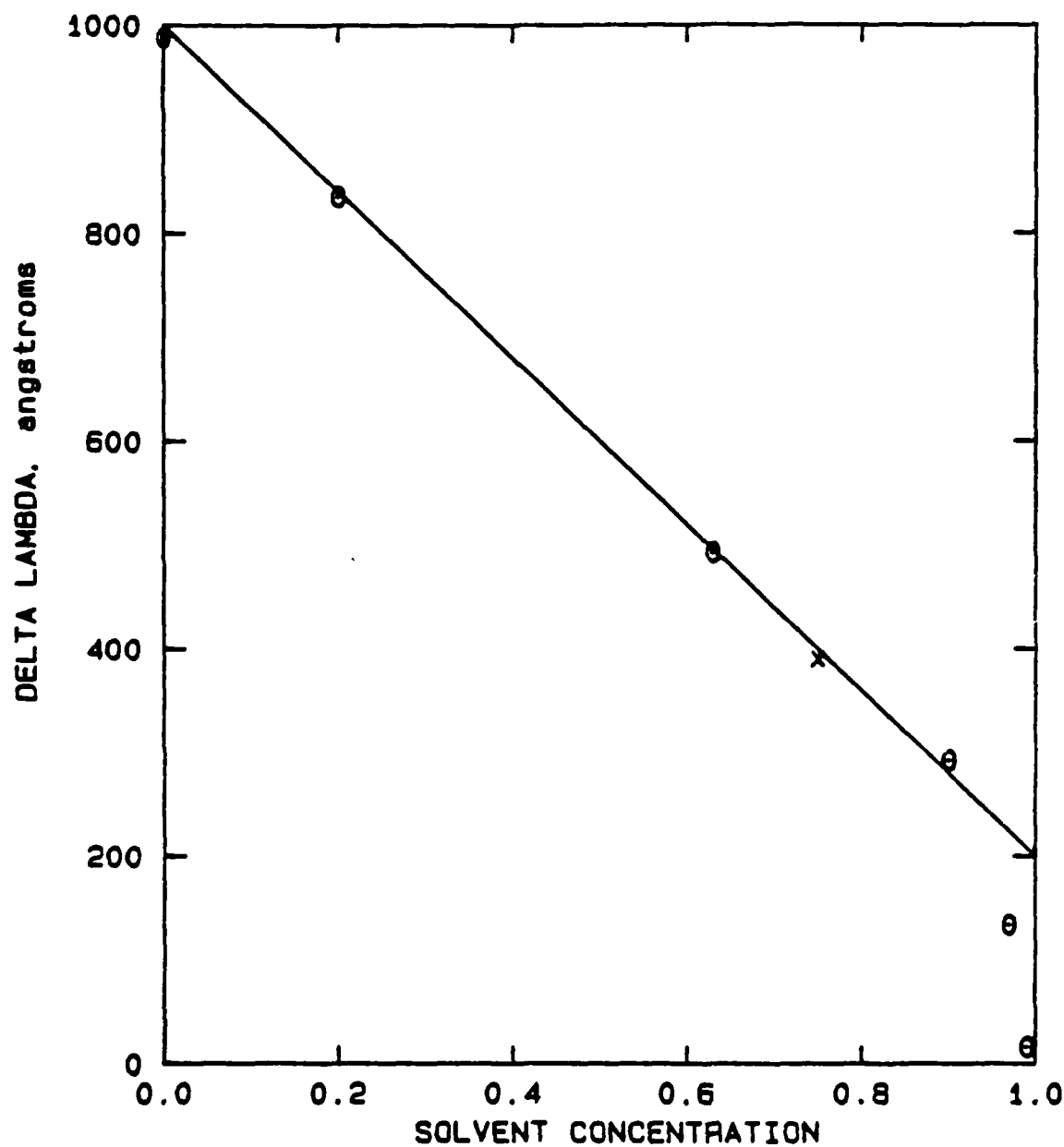


Fig. 8. Effect of dilution on band edge shift in CS_2 at 50 kbars. Solvent is ethanol or hexane.

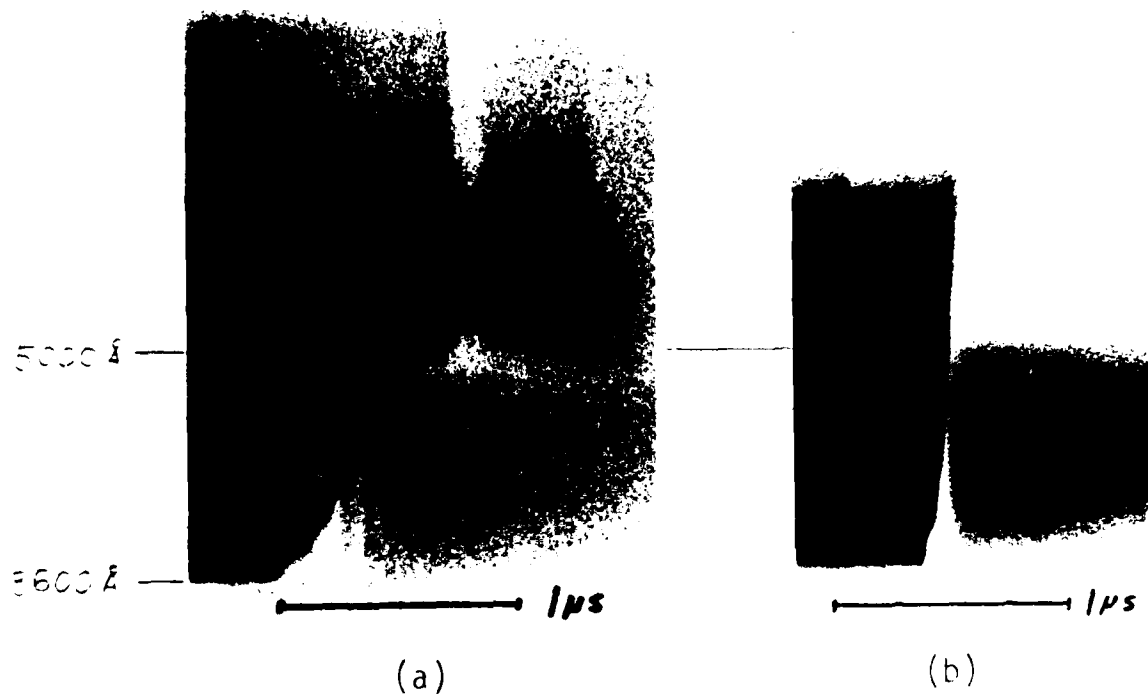


Fig. 9. Reflection experiments in CS_2 . SW loading.

(a) $P_f = 60 \text{ kb}$;

(b) $P_f = 91 \text{ kb}$.

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